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Dielectric Constant Decrease upon Illumination in a Barium Titanate Crystal

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Dielectric Constant Decrease upon Illumination in a Barium Titanate Crystal

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Abstract

A photodielectric effect has been observed in the *c*-axis dielectric constant of a barium titanate crystal in which illumination produces a decrease in the dielectric constant. When the crystal is exposed to illumination, the dielectric constant decreases in proportion to the time interval of the exposure, until a saturation level decrease of about 0.5 percent is reached. The rate of decrease is wavelength dependent. When illuminated with 403-nm light, the decrease has a value of -0.11 percent per incident millijoule per centimeter squared. The new constant level reached after saturation relaxes back toward the initial higher level when the illumination is removed. There is a partially irreversible component. Measurements made as a function of wavelength indicate that the decrease, before saturation, is proportional to total photo-carrier density. It is suggested that this photodielectric phenomenon could find applications in devices involving the optical control of dielectric properties.

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1. Introduction

The ability to dynamically modulate the dielectric constant of an insulating crystal using light is of interest because of possible applications to the optical control of millimeter-wave signals. Such applications include optically induced shifts of delay-line parameters or of the resonant frequency of a dielectric-filled cavity. Shifting the stop-band frequencies of photonic bandgap crystals is still another possibility. Thus, I was particularly interested in an unusual photodielectric effect recently observed in a barium titanate crystal, where the application of light caused a decrease in the *c*-axis dielectric constant, especially since this decrease seemed to be the result of a nonthermal process.

I observed the phenomenon with time-resolved measurements made at 8 MHz. This frequency is much lower than that at which the envisioned devices would operate. I think it likely, however, that the results will carry over to the millimeter-wave frequency range. For high-dielectric-constant ferroelectrics, the dc dielectric constant is known to generally persist into the multigigahertz range [1]. The photodielectric effect observed seems likely to persist at the much higher microwave and millimeter-wave frequencies. That is, the real part of the complex dielectric constant (which is the quantity more typically measured at high frequencies), when measured at millimeter-wave frequencies, will decrease when the crystal is illuminated, in a manner similar to that observed at 8 MHz.

2. Experiment

The barium titanate crystals used in the measurements were single-domain rectangular parallelepipeds. The measurements were made with a Hewlett Packard 4294A Impedance Analyzer. This instrument produces measurements of paired dielectric parameters, such as parallel capacitance and ac resistance, at successive time intervals, and displays these values, thus providing a way of determining these parameters as a function of time. The maximum sweep speed was 0.1 s per 0.5-in. division over a 5-in. chart, in which 402 point measurements are made over the span. A four-terminal connection was used to measure capacitance between electrodes. The oscillator strength was 0.5 V. A mechanical shutter was used to control the onset and occultation of illumination.

Measurements were made at 8 MHz. This is a frequency sufficiently high so that only the basic ionic and electronic mechanism for dielectric response is engaged, and the effects of the electromechanical response in this, a piezoelectric material, are avoided. Measurements were also made at frequencies as high as 15 MHz, and these showed results similar to those made at 8 MHz. Large apparent shifts due to electromechanical resonances were observed, however, at lower frequencies.

Two illumination sources were used: the optical output of a mercury arc lamp, used with spectral filters, and the expanded beam of an argon laser.

With the former, the responses at the mercury line wavelengths of 365, 403, 436, and 456 were measured. With the argon laser, the responses at the argon laser wavelengths of 454, 457, 465, 472, 476, 488, 496, 503, and 514 nm were determined.

When measuring the change of capacitance of a barium titanate capacitor with the application of light, one must take into account the strong dependence on temperature of the dielectric constant of the barium titanate ferroelectric. To eliminate the effects of the heating of the crystal, due to absorption of the incident light, I made the measurements at low-level illumination intensities, or alternatively, used only short illumination times. The effects of heating clearly appear with high-intensity illumination and with the application of illumination for more extended times. In these cases, there are significant temperature rises, and the response to illumination involves both the photodielectric effect and the thermal effect.

2.1 Photodielectric Shift with Incident Illumination as a Function of Illuminating Wavelength

Figure 1 shows the photo-induced change in the capacitance as a function of time, for a $5 \times 5 \times 2$ mm crystal with edges oriented along the crystal axes. Electrodes were perpendicular to the c -axis. A 5×5 mm face, unobscured by electrodes, was uniformly illuminated with 1.2 mW/cm^2 of 403-nm wavelength illumination. The illumination was produced by spectral filtering of the light from a high-pressure mercury arc lamp with a narrow-band interference filter. When the shutter is opened, the capacitance begins to decrease. The rate of decrease is linear with time and thus proportional to the incident integrated illumination flux. The capacitance change, from figure 1, is $-0.11 \text{ \%}/\text{mJ/cm}^2$. This change is assumed to be the result of a change in the bulk dielectric constant only; thus, photo-capacitive effects resulting from, for example, the effect of photoconductivity removing a blocking layer, are not a factor in the measurements. Under these assumptions, the percentage change in the dielectric constant is the same as that for the capacitance, that is, $-0.11 \text{ \%}/\text{mJ/cm}^2$. Figure 2 shows the response to 436-nm illumination measured in a similar way. The percentage change is $-0.016 \text{ \%}/\text{mJ/cm}^2$. At 454 nm, the percentage change is $-0.009 \text{ \%}/\text{mJ/mW/cm}^2$. A measurement was also made at 365-nm illumination, a wavelength that is well below that of the absorption edge. At this wavelength, the illumination does not penetrate the crystal but is absorbed near the surface. In this case, a small positive shift was seen.

At wavelengths above that of the absorption edge, where the bulk of the crystal is more or less uniformly illuminated, the dielectric constant decreases upon illumination, as we have seen, in proportion to the time the illumination is incident and thus in proportion to the time-integrated illumination flux. The percentage decrease per millijoule per centimeter squared is wavelength dependent, decreasing with increasing wavelength. Figure 3 plots the percentage shift per integrated flux unit as a function of wavelength for the above-band-edge wavelengths where the illumination is more or less uniformly absorbed. I have also plotted the measured

Figure 1. Change in c -axis capacitance upon illumination at 403 nm; illumination flux is 2 mW/cm^2 ; horizontal divisions, 200 ms; vertical divisions, 1 fF. Measured at 8 MHz with a 0.5-V rms field. Initial capacitance was 3.7 pF.

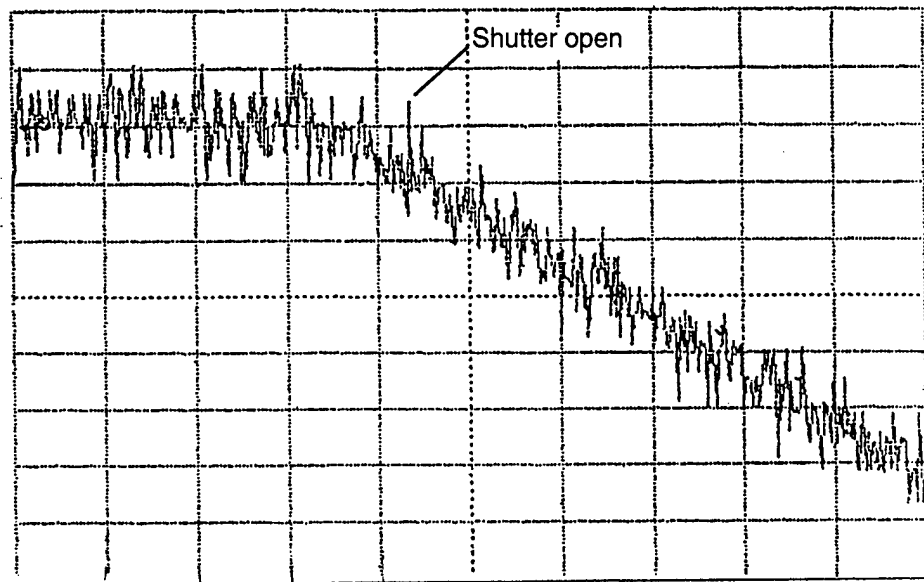
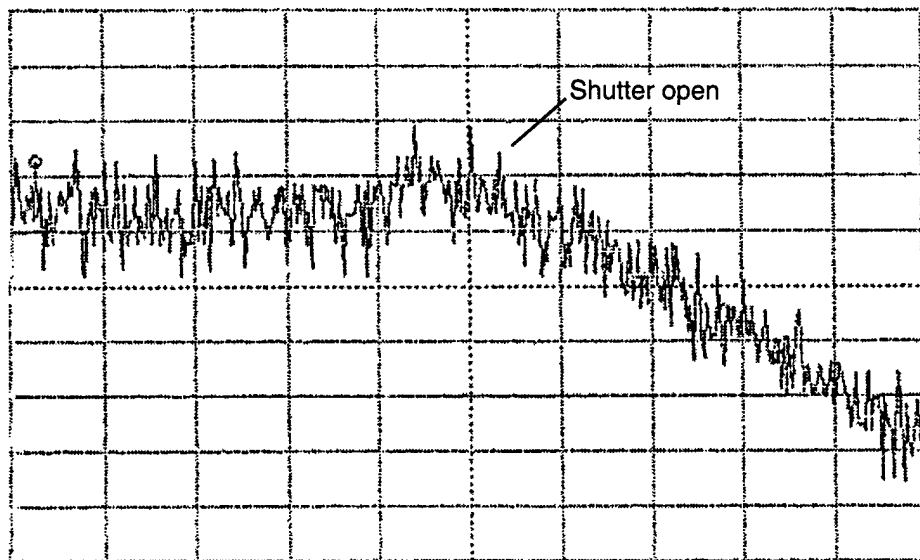


Figure 2. Change in c -axis capacitance upon illumination at 436 nm; illumination flux is 8.3 mW/cm^2 ; horizontal divisions, 200 ms; vertical divisions, 1 fF. Measured at 8 MHz with a 0.5-V rms field. Initial capacitance was 3.7 pF.



absorption coefficient as a function of wavelength. Note that the curves have a similar form. It is reasonable to assume that the change in dielectric constant proportional to the incident integrated flux results from the light flux absorbed. Since absorption in the visible in barium titanate is due to the creation of free carriers (photocarriers) [2], the similarity in the form of the two curves implies a percentage shift in dielectric constant proportional to total photocarrier density, the latter increasing in proportion to the absorbed illumination when carrier concentration is low.

Measurements were also made of dielectric constant changes produced by illumination from an argon ion laser at low intensities. Figure 4 shows the change in capacitance produced by a 200-ms light pulse, as a function of

Figure 3. Percentage decrease in dielectric constant per milliwatt per square centimeter as a function of illumination wavelength. Also shown is absorption coefficient as a function of wavelength.

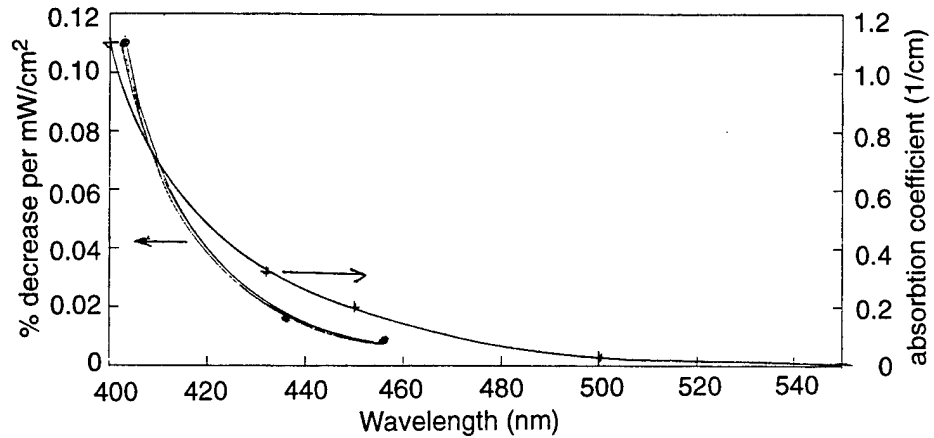
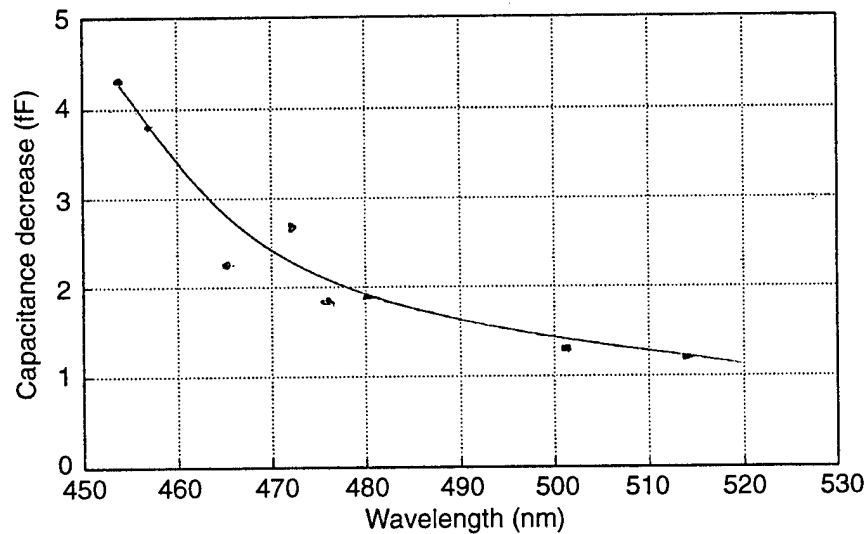


Figure 4. Decrease in *c*-axis capacitance of a barium titanate crystal as a function of wavelength resulting from 200-ms constant-intensity illumination pulses. Measurement was 8 MHz with a 0.5-V rms field. Initial capacitance was 3.7 pF.



wavelength. The dielectric constant decreases, as expected, and this decrease diminishes as the wavelength of the light gets longer; this change is consistent with decreased photocarrier density resulting from the longer wavelengths.

2.2 Saturation and Relaxation

The shift in dielectric constant, initially proportional to integrated flux, saturates. Figure 5 shows the response of the dielectric constant to illumination produced by opening and closing the shutter on a time scale long compared to that in the first figures. The illumination wavelength was 403 nm. The decrease saturates rather abruptly at about 0.5 percent. When the shutter is closed, the dielectric constant relaxes back toward its initial value, but not completely. There is an apparently irreversible component, although this, I observed, eventually vanishes over many minutes. Figure 6 shows the response at 436 nm. The results are similar to those in figure 5. We see, however, saturation occurring at a somewhat lower level, despite a higher illumination intensity.

Figure 5. Change in capacitance upon illumination and removal of illumination. Illumination wavelength is 403 nm, and illumination flux is 1.2 mW/cm^2 . Horizontal divisions, 1 min; vertical divisions, 2 fF.

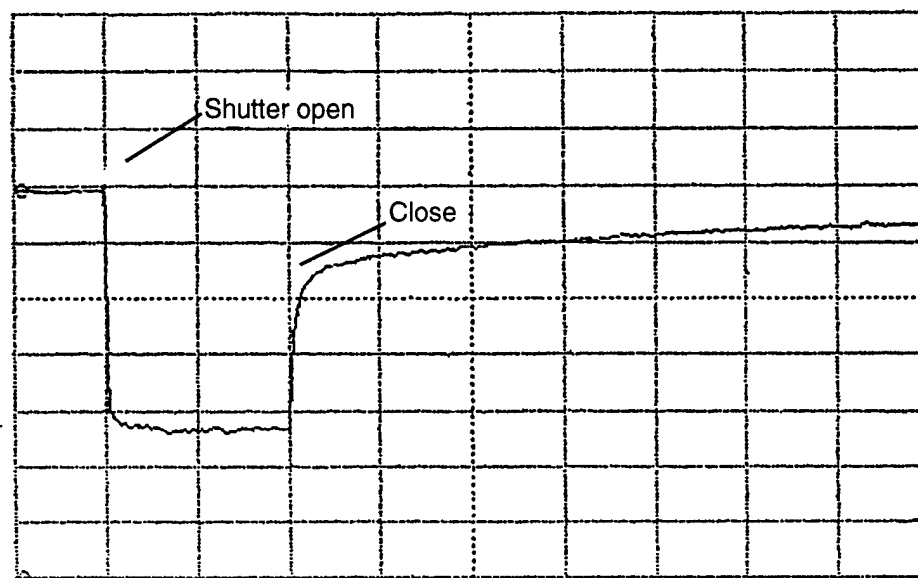
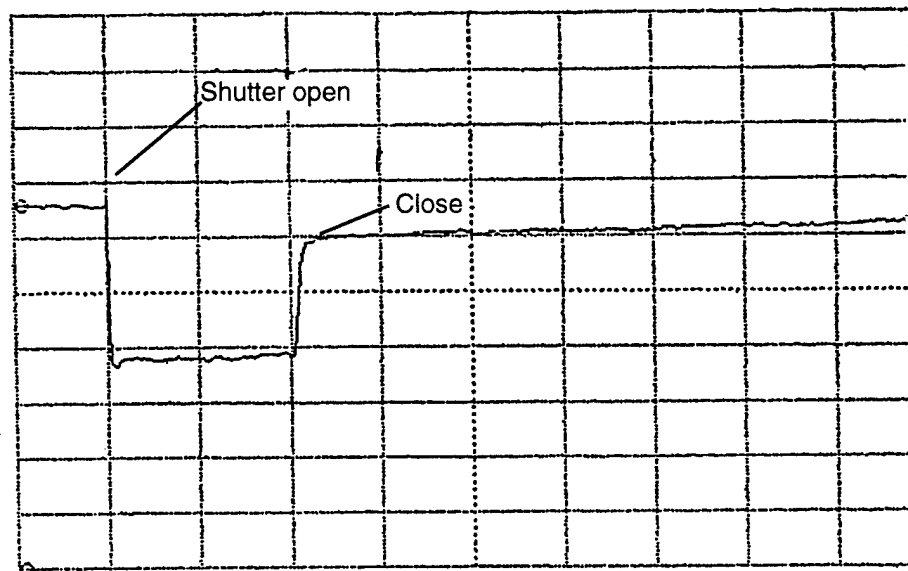


Figure 6. Same as in figure 5, but with an illumination flux of 8.3 mW/cm^2 at a wavelength of 436 nm.



2.3 Thermal Effects

Starting at room temperature, the *c*-axis dielectric constant increases monotonically with increasing temperature, rising to an anomalous peak at the approximately 120°C Curie transition temperature [3]. Thus, at room temperature the heating effects of illumination will generate a *c*-axis dielectric constant increase. This change is in a direction opposite to that observed for the photodielectric effect, where the application of illumination produces a decrease in the dielectric constant. The effects of the temperature produced by illumination-generated heating can be seen in figure 7. Here high-intensity illumination was used (the 488-nm output of an argon ion laser at about 1 W/cm^2). The shutter was opened and then, 1 s later, closed. With illumination, the dielectric constant drops to a negative saturation level, the result of the photodielectric effect. Then the dielectric

constant begins to rise as a result of the crystal heating. When the shutter is closed at 1 s, the crystal cools, and the dielectric constant decreases. At the same time, there is a continued negative photodielectric component that continues to decay. The dielectric constant's rise above its initial value is a result of the increment of temperature from the light pulse.

2.4 Change in ac Resistance

I also observed an increase in the ac resistance upon illumination, which then decays when illumination is removed. The direction of the change is different, but it otherwise follows the dielectric constant change. Figure 8 shows the response of the ac resistivity and also the change in dielectric constant. A relatively high intensity of about 1 W/cm^2 was used, but with a relatively short illumination time of 200 ms, so that the photodielectric

Figure 7. Response to high-intensity illumination of 1 W/cm^2 of 488-nm wavelength. Shutter is opened and then closed 1 s later. Figure shows effect of light-induced heating increasing dielectric constant. Horizontal divisions, 3 s; vertical divisions, 2 fF. Initial capacitance is 3.7 pF.

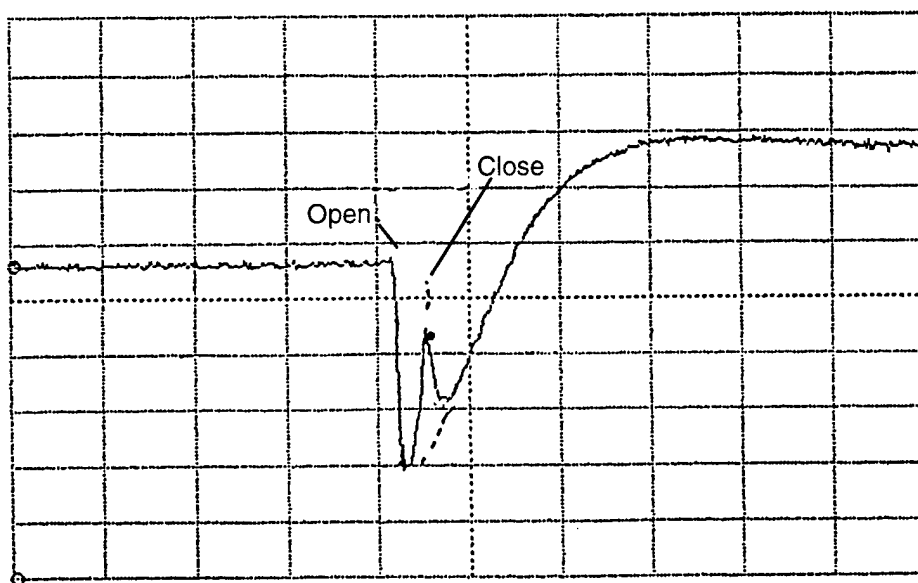
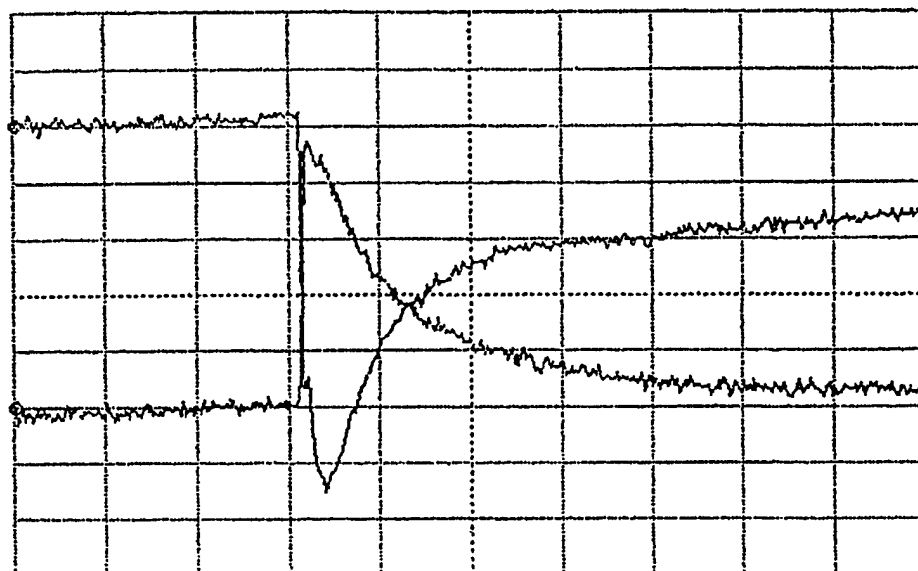


Figure 8. Response of *c*-axis capacitance and ac resistance to a 488-nm illumination pulse (note that they move in opposite directions). Horizontal divisions, 3 s; vertical divisions, 1 fF (capacitance) and 500 W (ac resistance). Initial capacitance is 3.7 pF. Initial ac resistance is 103,000 Ω .



effect predominates over the thermal effect. In the dielectric constant, we see the characteristic downward shift followed by the saturation of the shift and the characteristic relaxation; we also see a slowly decaying "irreversible" portion. We see just the opposite in the ac resistivity. It increases when the crystal is illuminated; the increase then relaxes back toward the initial level.

One peculiarity in the dielectric constant versus time curve in figure 8 is a continued decrease in the dielectric constant for a short time, preceding the relaxation after the termination of the illumination pulse.

2.5 Electrodes Perpendicular to *a*-Axis

A response to illumination was observed in the *a*-axis dielectric constant. The magnitude of the *a*-axis response was weak compared to that of the *c*-axis; it is presently not clear whether a significant photocarrier-induced response exists in the *a*-axis dielectric constant. A negative shift is seen at high intensities, but this is consistent with the initial effects of heating for the *a*-axis dielectric constant when it occurs at room temperature. The *a*-axis dielectric constant at room temperature decreases with increasing temperature [3]. Thus, one expects a negative thermally induced shift in the dielectric constant due to heating (in addition to any photodielectric effect). This decrease with increasing temperature changes to an increase with increasing temperature as the slope of the dielectric constant versus temperature curve changes at a temperature above room temperature but below the Curie transition temperature. The dielectric constant then again rises to an anomalous peak at the Curie temperature. Because of the nature of the thermal effect, the data for the *a*-axis case were difficult to interpret and I do not include these data here. However, the *a*-axis response appeared independent of wavelength, as one might expect for a thermal phenomenon.

3. Discussion

There is little in the literature with respect to photodielectric effects in barium titanate. A shift in the Curie temperature with illumination is described in Fridkin's book on photoferroelectrics [4]. Photodielectric effects are, however, found in other materials; for example, a photodielectric effect has been found in the ferroelectric calcium pyroniobate [4], which also includes a degree of irreversibility.

Characteristics of the *c*-axis photodielectric effect in barium titanate found in my experiments follow:

1. The rate of shift with exposure is proportional to the generated photocarrier concentration.
2. The change is a decrease linear with integrated illumination flux until it saturates.

3. There is an associated increase in the ac resistance.
4. When the illumination is blocked, there is a relaxation of the dielectric constant back toward its original level. The decay can be slow compared to the rise. This reversal is not complete; there is an irreversible portion. This may eventually decay, but at a rate that is slow on the time scale of these measurements.

The assumed equivalent circuit [6] treats the crystal as a pure, lossless capacitor of reactance

$$X = \frac{1}{\omega \epsilon' C_0} ,$$

in parallel with a resistor of resistance,

$$R_p = \frac{1}{\omega \epsilon'' C_0} ,$$

where ϵ' is the real part of the relative permittivity, ϵ'' is the imaginary part, and C_0 is the free space capacitance between the electrodes. The increase of R_p at a fixed frequency can thus be expected to be, as is the decrease in the real part, the result of a decrease in the imaginary part of the dielectric constant. This decrease is, in proportion to the density of photocarriers. Since the crystal is essentially insulating until illuminated, the photocarrier density totally constitutes the density of free carriers.

In a note from A. K. Jonscher, transmitted to me by E. Poindexter, Professor Jonscher suggests that the observed decrease in the dielectric permittivity of the barium titanate upon illumination may be the result of the dielectric permittivity of the free carriers. In this view, the free carriers are generated in proportion to the light intensity, with a response time that in the presence of intensive trapping exceeds the carrier lifetime by many orders of magnitude. The dielectric response of the carriers is obtained from a damped harmonic oscillator model assumption in the limiting case of inertia but no restoring force [7]. The free carrier density is then seen to contribute a negative component to both the real and the imaginary components of the dielectric constant. Such a decrease is consistent with the experimental observations in which the dielectric constant decreases and the ac resistivity increases upon illumination in proportion to the light-generated carrier density.

The consistency holds for the reversible portion of the change. There is no simple explanation for the irreversible portion, except that it would seem related to the filling of deep traps, which may change the dielectric properties by some other mechanism.

Another problem is the apparent difference in the *c*-axis and *a*-axis dielectric properties. Note that there is a difference in carrier mobility along these axes of about 2, with carriers moving along the *a*-axis having greater mobility at room temperature [8].

A detailed study comparing the predictions of the model with experiments is needed to verify Jonscher's suggestion; additional experimental measurements, particularly as a function of temperature, would provide valuable clues to its validity.

Jonscher points out, however, that if the suggestion is correct, there is then no need for ferroelectrics, and there is much wider scope for choice of materials: "you need a good semi-insulator with plenty of trapping and a high mobility ..."

The explanation if correct is also attractive from the point of view of microwave applications, for "the effect should continue into the high GHz frequency range ... the relevant frequency being $1/\tau_c = 100$ GHz."

4. Potential Applications

The shift with illumination due to the photodielectric effect is energy efficient when compared to shifts generated by the thermal effect. As a result, there may be applications in which a relatively low-intensity laser beam is used to control the operation of a device by changing the dielectric constant of a component dielectric element. If the effect that I have characterized at 8 MHz persists at the higher millimeter-wave frequencies (as expected), there could be valuable millimeter wave applications.

For example, illumination could be used to produce phase delays in millimeter-wave strip delay lines with a barium titanate crystalline dielectric. Optical control of such lines would be particularly useful in phased-array radar. Since quasi-single-crystal dielectric layers of *c*-axis barium titanate have been produced by laser deposition, the fabrication of such a delay line is a practical possibility.

Another possible application is the optical control of the stop-band frequencies of photonic bandgap (PBG) crystals. PBG crystals are three-dimensional (3-D) periodic structures of two interspersed dielectrics. A theoretical understanding of such structures shows the existence of frequency bands (band gaps) in which optical wave propagation is prohibited. These stop-band frequencies depend on the ratio of dielectric constant of the interspersed dielectrics. Because of this dependence, modulating the dielectric constant of either component should result in a modulation of the stop-band frequencies. Optical modulation of stop-band frequencies of PBG crystals could be used for electronic shutters and protective devices. Saturation shifts of even less than 0.5 percent, though relatively low, could still produce significant changes in the transmission band of a simple narrow-band millimeter-wave stack filter. Such modulation could be used to shutter a millimeter-wave channel for either protective or communication purposes.

5. Conclusion

I conclude that the decrease in the c -axis dielectric constant (measured at 8 MHz) of a barium titanate crystal in response to illumination is the result of the increase of photocarrier density. The effect is energy efficient where the photocarrier density is high, but the material is still transparent at wavelengths just above that of the absorption edge. The dielectric constant decrease with illumination saturated in these measurements at about 0.5 percent. This decrease is accompanied by an increase in ac resistivity. These quantities relax toward their original levels when illumination is removed. There remains, however, a smaller quasi-irreversible component, which may then decay further, but do so only slowly. The effect is not presently understood; however, the results that imply that a negative contribution to the real and imaginary part of the dielectric permittivity proportional to free carrier density is qualitatively consistent with a model [7] describing the contribution of free carriers with collisions to the complex dielectric permittivity.

The model also predicts that the photodielectric effect (characterized here at the relatively low frequency of 8 MHz) will persist into the much higher millimeter-wave frequencies; if so, interesting millimeter-wave applications are possible. These involve the use of changes in dielectric constant of dielectric lines to vary phase velocity and also resonant cavity frequency. Also possible are applications involving the optical modulation of stop-band frequencies in millimeter-wave photonic bandgap crystals.

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